

PATENT COOPERATION TREATY

From the INTERNATIONAL BUREAU

PCT

NOTIFICATION OF ELECTION

(PCT Rule 61.2)

To:

Commissioner
US Department of Commerce
United States Patent and Trademark
Office, PCT
2011 South Clark Place Room
CP2/5C24
Arlington, VA 22202
ETATS-UNIS D'AMERIQUE

in its capacity as elected Office

Date of mailing (day/month/year) 30 October 2000 (30.10.00)	Applicant's or agent's file reference 49513
International application No. PCT/FI00/00098	Priority date (day/month/year) 12 February 1999 (12.02.99)
International filing date (day/month/year) 11 February 2000 (11.02.00)	
Applicant ALASTALO, Kauno et al	

1. The designated Office is hereby notified of its election made:

☒ in the demand filed with the International Preliminary Examining Authority on:
05 September 2000 (05.09.00)

☐ in a notice effecting later election filed with the International Bureau on:

2. The election ☒ was

☐ was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

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The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland	Authorized officer Charlotte ENGER
Facsimile No.: (41-22) 740.14.35	Telephone No.: (41-22) 338.83.38

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From the INTERNATIONAL BUREAU

NOTIFICATION OF THE RECORDING OF A CHANGE

(PCT Rule 92bis.1 and
Administrative Instructions, Section 422)

To:

BERGGREN OY AB
P.O. Box 16
FIN-00101 Helsinki
FINLANDE

Date of mailing (day/month/year)
13 October 2000 (13.10.00)

Applicant's or agent's file reference
49513

International application No.
PCT/FI00/00098

International filing date (day/month/year)
11 February 2000 (11.02.00)

IMPORTANT NOTIFICATION

1. The following indications appeared on record concerning:

☒ the applicant ☐ the inventor ☐ the agent ☐ the common representative

Name and Address

BOREALIS A/S
Lyngby Hovedgade 96
DK-2800 Lyngby
Denmark

State of Nationality

DK

State of Residence

DK

Telephone No.

Facsimile No.

Teleprinter No.

2. The International Bureau hereby notifies the applicant that the following change has been recorded concerning:

☒ the person ☐ the name ☐ the address ☐ the nationality ☐ the residence

Name and Address

BOREALIS TECHNOLOGY OY
P.O. Box 330
FIN-06101 Porvoo
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State of Nationality

FI

State of Residence

FI

Telephone No.

Facsimile No.

Teleprinter No.

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3. Further observations, if necessary:

4. A copy of this notification has been sent to:

<input checked="" type="checkbox"/> the receiving Office	<input checked="" type="checkbox"/> the designated Offices concerned
<input type="checkbox"/> the International Searching Authority	<input type="checkbox"/> the elected Offices concerned
<input type="checkbox"/> the International Preliminary Examining Authority	<input type="checkbox"/> other:

The International Bureau of WIPO
34, chemin des Colombettes
1211 Geneva 20, Switzerland

Facsimile No.: (41-22) 740.14.35

Authorized officer



Aino Metcalfe

Telephone No.: (41-22) 338.83.38

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INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference 49513	FOR FURTHER ACTION See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/FI00/00098	International filing date (day/month/year) 11/02/2000	Priority date (day/month/year) 12/02/1999
International Patent Classification (IPC) or national classification and IPC C08F4/44		
Applicant BOREALIS TECHNOLOGY OY		
<p>1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.</p> <p>2. This REPORT consists of a total of 5 sheets, including this cover sheet.</p> <p><input type="checkbox"/> This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).</p> <p>These annexes consist of a total of sheets.</p>		
<p>3. This report contains indications relating to the following items:</p> <p>I <input checked="" type="checkbox"/> Basis of the report</p> <p>II <input type="checkbox"/> Priority</p> <p>III <input type="checkbox"/> Non-establishment of opinion with regard to novelty, inventive step and industrial applicability</p> <p>IV <input type="checkbox"/> Lack of unity of invention</p> <p>V <input checked="" type="checkbox"/> Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement</p> <p>VI <input type="checkbox"/> Certain documents cited</p> <p>VII <input type="checkbox"/> Certain defects in the international application</p> <p>VIII <input checked="" type="checkbox"/> Certain observations on the international application</p>		
Date of submission of the demand 05/09/2000	Date of completion of this report 20.04.2001	
Name and mailing address of the international preliminary examining authority:  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer Adams, F Telephone No. +49 89 2399 8511 	

**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT**

International application No. PCT/FI00/00098

I. Basis of the report

1. With regard to the **elements** of the international application (*Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17)*):

Description, pages:

1-15 as originally filed

Claims, No.:

1-27 as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☐ the claims, Nos.:
- ☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/FI00/00098

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims	1-27
	No:	Claims	
Inventive step (IS)	Yes:	Claims	1-27
	No:	Claims	
Industrial applicability (IA)	Yes:	Claims	1-27
	No:	Claims	

2. Citations and explanations
see separate sheet

VIII. Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:
see separate sheet

Ad V:

- 1). The present application satisfies the criteria set forth in Article 33(2) and 33(3) PCT because the subject-matter of the claims is new and inventive in respect of prior art as defined in the regulations (Rule 64(1)-(3) PCT).

None of the cited documents discloses a catalyst system for the polymerization of alpha-olefins which is prepared by a first activation step comprising contacting a solid transition metal compound with an organoaluminum compound in the presence of an oil and contacting in a second step the obtained reaction mixture with an organoaluminum compound to obtain an activated catalyst, and a prepolymerization step comprising prepolymerizing a monomer in the presence of the activated catalyst, or makes such a catalyst system obvious.

- 2). EP-A-0588277 discloses the activation of a Ziegler-type catalyst by contacting the catalyst with TEAL in the presence of hexane and the prepolymerization of the activated catalyst with propene. The activated and prepolymerized catalyst is subsequently diluted with a mineral oil and used for propene polymerization in the presence of further TEAL. Nothing is said about the activation in the presence of mineral oil prior to the prepolymerization.

EP-A-0887379 discloses the polymerization of propene in the presence of a prepolymerized transition metal catalyst and an organoaluminum compound as an activating agent. The prepolymerization may be carried out in the presence of a viscous substance such as an olefinic wax. Nothing is said about an activation step in the presence of an oil.

US-A-5641721 (D1) discloses the activation of a titanium based catalyst with triethylaluminum in the presence of PAO oil and the prepolymerization of the thus obtained activated catalyst with propylene in the presence of further triethyl aluminum (cf. example 1; column 4, lines 39 to column 5, lines 54). The difference between the catalyst system as claimed and the catalyst system of D2 is that in D2 the procatalyst (i.e. the transition metal) and the cocatalyst, (i.e. the organoaluminum compound), are first combined and then added to the oil (see column 5, lines 18-22; column 7, lines 31-37).

EP-A-0573862 discloses the preparation of a supported titanium-based catalyst prepared by heating MgCl_2 , TiCl_4 and vaseline oil, the activation of the supported catalyst with TEAL and the subsequent prepolymerization of the catalyst with propene. Nothing is said about an activation step in the presence of an oil and TEAL.

Ad VIII:

- 1). The definition of the catalyst system comprising a "solid transition metal compound" is unclear (Art. 6 PCT). It appears from the description on page 5, line 21 and the examples that the transition metal of the invention is titanium. Alternatively, defining the transition metal as a metal belonging to the Groups 4 to 6 of the Periodic Table of Elements (page 1, lines 28-32) may also clarify claim 1.
- 2). The term "oil" used in claim 1 is vague and unclear and leaves the reader in doubt as to the meaning of the technical feature to which it refers, thereby rendering the definition of the subject-matter of said claim unclear (Article 6 PCT). It should be noted that it depends inter alia on the temperature whether a compound is an oil or a wax.
- 3). Unclear expressions like "about" and "and the like" were not deleted throughout the claims and the description (Art. 6 PCT).

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau



(43) International Publication Date
17 August 2000 (17.08.2000)

PCT

(10) International Publication Number
WO 00/47638 A3

- (51) International Patent Classification⁷: C08F 4/44, 10/00
- (21) International Application Number: PCT/FI00/00098
- (22) International Filing Date: 11 February 2000 (11.02.2000)
- (25) Filing Language: English
- (26) Publication Language: English
- (30) Priority Data:
990283 12 February 1999 (12.02.1999) FI
- (71) Applicant (for all designated States except US): **BOREALIS A/S** [DK/DK]; Lyngby Hovedgade 96, DK-2800 Lyngby (DK).
- (72) Inventors; and
- (75) Inventors/Applicants (for US only): **ALASTALO, Kauno** [FI/FI]; Karjapolku 4, FIN-06400 Porvoo (FI). **LESKINEN, Pauli** [FI/FI]; Kantakyläntie 6 D, FIN-00640 Helsinki (FI).
- (74) Agent: **BERGGREN OY AB**; P.O. Box 16, FIN-00101 Helsinki (FI).
- (81) Designated States (*national*): AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CR, CU, CZ, DE, DK, DM, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW.
- (84) Designated States (*regional*): ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).
- Published:**
— With international search report.
- (88) Date of publication of the international search report:
7 December 2000
- For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.*

(54) Title: AN α -OLEFIN POLYMERIZATION CATALYST SYSTEM AND ITS USE FOR THE POLYMERIZATION OF α -OLEFINS

(57) Abstract: In the invention, a catalyst system for the polymerization of α -olefins has been prepared by a process including a catalyst activation comprising the contacting of a solid transition metal compound with an organoaluminium compound, and a catalyst prepolymerization comprising the polymerization of a premonomer in the presence of the activated catalyst. High activity, low fines polymerization is accomplished if the catalyst activation comprises a first step of contacting the solid transition metal compound with a first organoaluminium compound in the presence of an oil to give a first reaction mixture, and a second step of contacting the first reaction mixture with a second organoaluminium compound to give a second reaction mixture, the second organoaluminium compound being the same as or different from the first organoaluminium compound.

WO 00/47638 A3

INTERNATIONAL SEARCH REPORT

International Application No.

PCT/FI 00/00098

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C08F4/44 C08F10/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C08F

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 588 277 A (FINA TECHNOLOGY) 23 March 1994 (1994-03-23) page 2, line 46 - line 50 page 3, line 15 - line 20 page 5, line 50 - page 6, column 7 ---	1-27
A	EP 0 887 379 A (BOREALIS AS) 30 December 1998 (1998-12-30) column 12, line 23 - line 28 ---	1-27
A	US 5 641 721 A (PENTTI ISMO ET AL) 24 June 1997 (1997-06-24) claims, abstract column 4, line 53 - line 63 --- -/--	1-27



Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

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"&" document member of the same patent family

Date of the actual completion of the international search

7 June 2000

Date of mailing of the international search report

04.08.2000

Name and mailing address of the ISA

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Authorized officer

HELENA DANIELSSON

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 0 573 862 A (HIMONT INC) 15 December 1993 (1993-12-15) page 4, line 4 - line 16 page 6, line 25 - line 37 claims -----	1-27

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No.

PCT/FI 00/00098

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP 0588277 A	23-03-1994	US 4767735 A	30-08-1988
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EP 0887379 A	30-12-1998	FI 972726 A	25-12-1998
		AU 7921098 A	04-01-1999
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		WO 9858975 A	30-12-1998
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US 5641721 A	24-06-1997	FI 925913 A	30-06-1994
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		RU 2081132 C	10-06-1997

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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification ⁷ : C08F 10/00	A2	(11) International Publication Number: WO 00/47638 (43) International Publication Date: 17 August 2000 (17.08.00)
(21) International Application Number: PCT/FI00/00098 (22) International Filing Date: 11 February 2000 (11.02.00) (30) Priority Data: 990283 12 February 1999 (12.02.99) FI (71) Applicant (for all designated States except US): BOREALIS A/S [DK/DK]; Lyngby Hovedgade 96, DK-2800 Lyngby (DK). (72) Inventors; and (75) Inventors/Applicants (for US only): ALASTALO, Kauno [FI/FI]; Karjapolku 4, FIN-06400 Porvoo (FI). LESKINEN, Pauli [FI/FI]; Kantakyläntie 6 D, FIN-00640 Helsinki (FI). (74) Agent: BERGGREN OY AB; P.O. Box 16, FIN-00101 Helsinki (FI).		(81) Designated States: AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CR, CU, CZ, DE, DK, DM, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG). Published <i>Without international search report and to be republished upon receipt of that report.</i>
(54) Title: AN α -OLEFIN POLYMERIZATION CATALYST SYSTEM AND ITS USE FOR THE POLYMERIZATION OF α -OLEFINS		
(57) Abstract <p>In the invention, a catalyst system for the polymerization of α-olefins has been prepared by a process including a catalyst activation comprising the contacting of a solid transition metal compound with an organoaluminium compound, and a catalyst prepolymerization comprising the polymerization of a premonomer in the presence of the activated catalyst. High activity, low fines polymerization is accomplished if the catalyst activation comprises a first step of contacting the solid transition metal compound with a first organoaluminium compound in the presence of an oil to give a first reaction mixture, and a second step of contacting the first reaction mixture with a second organoaluminium compound to give a second reaction mixture, the second organoaluminium compound being the same as or different from the first organoaluminium compound.</p>		

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DK	Denmark	LR	Liberia	SG	Singapore		
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An α -olefin polymerization catalyst system and its use for the polymerization of α -olefins

5 The invention relates to a catalyst or system for the polymerization of α -olefins, the catalyst being prepared by a process including a catalyst activation comprising the contacting of a solid transition metal compound with an organoaluminium compound, and a catalyst prepolymerization comprising the polymerization of a premonomer in the presence of the activated catalyst.

10 The word "system" means a complex unity formed of many often diverse parts subject to a common plan or serving a common purpose. Thus, a catalyst system for the polymerization of olefins means a unity formed of diverse catalyst components serving the common purpose of polymerizing olefins. The present catalyst system may or may not include further components.

15 The word "including" means that the process may have other steps than said activation and prepolymerization steps, as well. The word "comprising" means that the activation and prepolymerization steps may have other operations than said contacting and polymerization, as well.

The invention also relates to a process for the polymerization of an α -olefin, wherein an α -olefin is contacted with a catalyst system of the above type.

20 By α -olefin monomer is in this connection meant an α -olefin which is capable of polymerization by the insertion (Ziegler-Natta) mechanism. An α -olefin is a compound having the structure $\text{CH}_2 = \text{CHR}$, wherein R is a linear or cyclic alkyl group. Typical α -olefin monomers of the invention are propene ($\text{R} = -\text{CH}_3$), butene-1 ($\text{R} = -\text{CH}_2\text{CH}_3$), 4-methylpentene-1 ($\text{R} = \text{CH}_2\text{CH}(\text{CH}_3)_2$), hexene-1
25 ($\text{R} = -(\text{CH}_2)_3\text{CH}_3$) and octene-1 ($\text{R} = -(\text{CH}_2)_5\text{CH}_3$). By an α -olefin polymer is meant an α -olefin homopolymer or copolymer. As monomers to be copolymerized can, in addition to α -olefin monomers of the above mentioned type, also be used ethene.

30 α -olefins alone or with other unsaturated monomers can often be polymerized in the presence of a so called Ziegler-Natta catalyst system, which has essentially two components: a catalyst (also called procatalyst) comprising a compound of a transition metal belonging to groups 4 to 6 of the Periodic Table of Elements (IUPAC 1990), and a cocatalyst based on an organic compound of a metal belonging to any of groups 1 to 3 and 13 of said Table. Typical compounds of

transition metals are the chlorides and especially the tetrachloride of titanium. Typical organometallic cocatalysts are organoaluminium compounds such as aluminium alkyl compounds and especially the trialkyl aluminiums. This kind of Ziegler-Natta catalyst system has been further developed by depositing and thus solidifying the transition metal compound on a more or less inert and particulate support and by adding to the catalyst composition in the stages of its preparation several additives, among others internal and external electron donors. A typical support is magnesium chloride, typical internal electron donors are the dialkyl phthalates and typical external electron donors are the alkyl alkoxy silanes. These compounds have improved the polymerization activity, the operating life and other properties of the catalyst system and above all the properties of the polymers which are obtained by means of the catalyst system. In order to further improve the properties of such a catalyst system at least a part of it has been contacted with a small amount of monomer to give a polymer coated, so called prepolymerized catalyst or catalyst system.

According to the prevailing conventional technology, the Ziegler-Natta catalyst is used in particulate form for the polymerization of olefins. Further, a precontacting thereof with a part of the cocatalyst has been used to improve the polymerization results. In order to facilitate catalyst dosing, the solid catalyst has often been combined with a wax, which enables a continuous feed thereof into the polymerization reactor. Instead of wax, a mixture thereof with oil has also been used. US 5,641,721 discloses a process, in which a solid transition metal compound is first precontacted with a first aluminium alkyl compound, the obtained contacting product is added to a polymerization reactor containing a second aluminium alkyl compound and a wax/oil mixture, and the resulting mixture is used for the prepolymerization of propene.

However, conventional polymerization catalyst systems have the disadvantage of producing too little polymer and/or too large a fraction of polymer having a very small particle size; so called fines. By amount of fine particles is meant the percentage by weight of polymer particles of defined diameter < 0.1 mm or < 0.25 mm, determined by sieving of the polymerization process outcome using sieves according to ASTM 1921. The problem is pronounced when producing high isotacticity polypropene having low catalyst residue and good processability (high melt flow rate). Among other inconveniences, the fines are accumulated in the gas recycling equipment of gas phase polymerization reactors, plugging filters and causing fouling on transportation lines.

The purpose of the invention is to provide a catalyst system for the polymerization of olefins, which gives olefin polymers of low catalyst residue, good processability and high stereoregularity in combination with a low amount of fines. This purpose has now been implemented by providing a new type of catalyst system for the polymerization of olefins. The catalyst system has been prepared by a process including a catalyst activation comprising the contacting of a solid transition metal compound with an organoaluminium compound, and a catalyst prepolymerization comprising the polymerization of a premonomer in the presence of the activated catalyst. The catalyst activation step comprises a first step of contacting the solid transition metal compound with a first organoaluminium compound in the presence of an oil to give a first reaction mixture, and a second step of contacting the first reaction mixture with a second organoaluminium compound to give a second reaction mixture, the second organoaluminium compound being the same as or different from the first organoaluminium compound. It is important that at least some oil is present when the solid transition metal compound and the first organoaluminium compound are contacted for the first time. The method described in this invention allows the reduction of the amount of fine particles (diameter < 0.25 mm) down to 0.5 w-% or below with no deterioration of other product or process characteristics, that is, for example the isotacticity, activity and molar mass of the product produced by this process are identical or improved compared to the traditional process.

Any order of addition may be used, as long as the oil is present when the solid transition metal catalyst compound and the first organoaluminium compound are contacted in the first step. According to a non-limiting explanation, contacting without medium or in low viscosity medium gives too vigorous a reaction which breaks the particles of the solid transition metal catalyst component. The deteriorated catalyst morphology is then reflected as an increased amount of fines in the polymer. On the other hand, using only wax, fat, paraffin or prepolymer when contacting prevents the contacting reaction almost completely and postpones it to the actual polymerization where the reaction will again be too vigorous and break the particles. By adding oil to the contacting of the first step, a smooth, suppressed reaction between the solid transition metal compound and the first organoaluminium compound is accomplished. The aim of the invention is to reach a mild activation of the solid transition metal compound by the organometal compound. The addition of oil allows the reaction which is a reduction of titanium from Ti^{4+} to Ti^{3+} to proceed slowly. Good morphology is thus obtained and the carrier's pores are more open

since they are not filled with wax. A longer lifetime for the catalyst system and consequently a more economic process is reached.

According to one embodiment of the invention, the sequence of addition in the first step is such that both the oil and a wax, fat, solid paraffin or the like are present when contacting the solid transition metal catalyst compound and the first organo-
5 aluminium compound. If the first organoaluminium compound is called A, the solid transition metal compound is called C, the wax, fat, solid paraffin or the like is called W and the oil is called O, then this embodiment is exemplified by the following basic sequences of addition: (A + O) + (C + W), (A + W) + (C + O),
10 [(A + O) + W] + C, [A + (O + W)] + C, and [(A + W) + O] + C, A + [(C + O) + W], A + [C + (O + W)], and A + [(C + W) + O]. Additionally, small amounts of each component may be freely distributed over the other components in order to optimize the effect of the addition sequence. In this embodiment, the contacting reaction is smoothened by a compatible mixture of both the oil and the wax, fat,
15 solid paraffin or the like, and is suitable for contacting reactions which would otherwise have been exceptionally vigorous.

However, only oil, and not, for example, wax, fat, solid paraffin or the like, is preferably present when contacting the solid transition metal compound and the first organoaluminium compound in the first step. If the wax, fat, solid paraffin and the
20 like is added later in said first step this embodiment is exemplified by the following basic sequences of addition: [(A + O) + C] + W and [A + (C + O)] + W. These sequences can be modified by adding amounts of each component to any other component, providing that wax, fat, solid paraffin or the like is not present in the contacting. Thus, among others, the sequences: [(A + O) + (C + O)] + W,
25 [(A + O) + C] + (W + O) and [A + (C + O)] + (W + O) are obtained. Further, instead of O, a mixture of a major amount of O and a minor or very small amount of W may be used, to smoothen, suppress or retard the reaction in a regulated way during the first step.

Most preferably, in the contacting of said first step, a mixture consisting essentially
30 of the solid transition metal compound and the oil is contacted with the first organoaluminium compound. This corresponds to the above basic sequence A + (C + O).

In the contacting of the first step, the weight ratio between the solid transition metal compound and the oil is preferably between 0.1 and 5, more preferably between 0.2
35 and 1, most preferably between 0.3 and 0.8. Said mixture consisting essentially of

the solid transition metal compound and the oil has preferably been prepared by heating them together at an elevated temperature, preferably at a temperature between about 26 °C and about 100 °C, most preferably at a temperature between about 30 °C and about 80 °C.

5 As was stated above, it is essential to prevent the contacting reaction between the solid transition metal compound and the first organoaluminium compound from becoming too vigorous and breaking the particles of the the solid transition metal catalyst component. According to the main embodiment of the invention, this is accomplished by using oil in the first step of the activation. A further means of
10 strengthening the retarding or suppressing effect of the oil on the reaction, is the use of low temperatures. Thus, during the contacting of said first step, the solid transition metal compound, the first organoaluminium compound and the oil are preferably precontacted at a lowered temperature, more preferably at a temperature between about -20 °C and about +20 °C, most preferably at a temperature between
15 about 0 °C and about +16 °C.

The intensity of the reaction between the solid transition metal catalyst component and the organoaluminium cocatalyst also depends on their ratio. Preferably, in the contacting of said first step, the first organoaluminium (Al_1) compound and the solid transition metal (Tr) compound are contacted in the presence of the oil in an atomic
20 ratio Al_1/Tr of between about 0.5 and about 5, preferably between about 1 and about 3. Usually, Tr is titanium Ti. Further, the contacting reaction proceeds at a certain rate, meaning that a certain reaction time is needed to obtain a desired yield of reaction product which usually is reduced titanium tetrachloride. Preferably, the solid transition metal compound, the first organoaluminium compound and the oil
25 are precontacted for a time period enough to allow said first organoaluminium compound and said solid transition metal compound to react completely, more preferably for about 10 min to about 60 h and most preferably for about 10 h to 24 h.

According to one embodiment of the invention, this step is performed as a batch
30 step separately from the polymerization process.

As was said before, the wax, the fat, the solid paraffin or the like may be, but preferably is not, present in the contacting the first of step. When the wax, fat, solid paraffin or the like is not present said initial contacting, it may be added to the formed first reaction mixture immediately after the initial contacting or it may later
35 be combined with the second organoaluminium cocatalyst of the second step or

added with the prepolymerization reactants. Preferably, it is added immediately after the initial contacting of the solid transition metal compound, the first organoaluminium compound and the oil. Thereby, the contacted solid transition metal compound, the organoaluminium compound and the oil are contacted directly
5 with the wax, fat, solid paraffin or the like to give a waxed first reaction mixture.

According to one embodiment of the invention, the wax, fat, solid paraffin or the like is used alone when contacting it with the first reaction mixture. According to another embodiment, the wax, fat, solid paraffin or the like is added in the form of a mixture of said wax, fat, solid paraffin or the like and an oil. The oil of the mixture
10 is preferably the same oil which is present during the initial contacting of the solid transition metal compound and the organoaluminium compound.

In the first step, said wax, fat, solid paraffin or the like preferably has an elevated temperature, where it is in a molten state. More preferably the elevated temperature is between about 60 °C and about 160 °C, most preferably between about 80 °C and
15 about 140 °C. On the other hand, the temperature of the first reaction mixture is during contacting with the wax, fat, solid paraffin or the like below about 60 °C, preferably below about 50 °C, most preferably below about 40 °C.

When adding the wax, fat, solid paraffin or the like alone and directly to the first reaction mixture, it is preferable, if, after the addition, the temperature is lowered to
20 below about 30 °C, more preferably below about 20 °C and most preferably below about 15 °C. This lowering of the temperature thus takes place at the end of the first step before performing the step of contacting the product of the second step with the second organoaluminium compound.

In the second step of said activation, which the non-waxed first reaction mixture is
25 contacted with a second organoaluminium compound. If the non waxed first reaction product is contacted in the second step, said wax, fat, solid paraffin or the like is optionally added at some later stage. Preferably, the first reaction mixture is contacted both with the second organoaluminium compound and an external electron donor. Further catalyst components may be also added in the second step.
30 Usually, the contacting of the second step takes place in the prepolymerization reactor, preferably immediately before starting the actual prepolymerization reaction. The second step of activation needs a very short time, preferably only a few minutes, most preferably only 1 to 3 minutes, whereby the whole polymerization process time shortens.

As a whole, the catalyst system of the invention has preferably been prepared by a process in the first activation step of which a mixture consisting essentially of the solid transition metal catalyst compound and the oil is precontacted with the first organoaluminium compound to give a first reaction mixture. Then, the first reaction mixture is contacted with the wax, fat, solid paraffin or the like and optionally the remaining part of the oil, to give the waxed first reaction mixture. After that, the waxed first reaction mixture is in a second activation step contacted with a second organoaluminium compound to give a second reaction mixture. Finally, an olefin premonomer is prepolymerized in the presence of at least the second reaction mixture, to give said prepolymerizate.

If a mixture of oil and wax, fat, solid paraffin or the like, is used, the weight ratio between the total amount of said oil and the total amount of said wax, fat, solid paraffin or the like is such that the viscosity of their mixture at 20-25 °C is about 1 Pa·s to about 15 Pa·s, preferably about 4 Pa·s to about 10 Pa·s.

- 15 The atomic ratio between the aluminium (Al_1) of said first organoaluminium compound and the aluminium (Al_2) of said second organoaluminium compound Al_1/Al_2 is preferably between about 0.001 and about 1, most preferably between about 0.01 and about 0.1. Further, the atomic ratio between the aluminium (Al) of the total amount of organoaluminium compounds and the transition metal (Tr) of the solid transition metal compound depends on whether the catalyst system is used for polymerization alone or together with an additional, third organoaluminium compound. Anyhow, the total Al/Tr is preferably between about 10 and about 1000, most preferably between about 50 and about 500. The external electron donor acts during the polymerization of C_3 - and higher α -olefins as a stereoregulating agent.
- 20
- 25 The atomic to molar ratio between the aluminium (Al) of the total amount of aluminium compounds and the amount of external electron donor ED is preferably between about 1 and about 100, most preferably between about 10 and about 50.

In the invention, a solid transition metal compound is used. By definition the solid transition metal compound of the invention is a solid catalyst or catalyst component comprising a transition metal compound which is catalytically active in the polymerization of olefins and is solid and/or supported on a solid substance. Preferably it has been prepared by contacting at least magnesium dichloride or a complex thereof, titanium tetrachloride and an internal electron donor.

30

35 In order to act as support for the titanium tetrachloride and the internal electron donor, the magnesium dichloride must be in a chemically active form. This means

that the magnesium dichloride used in the claimed process must have lower crystallinity and higher specific surface area than commercial magnesium dichloride.

5 Magnesium dichloride may be activated mechanically e.g. by dry-comilling anhydrous magnesium dichloride with an internal electron donor. Thereafter, the comilled product is heat-treated with an excess of titanium tetrachloride, followed by repeated washings with titanium tetrachloride and/or hydrocarbons. Typically, such catalyst components exhibit a high specific surface area (50-300 m²/g) and contain from 0.5 to 3% by weight of titanium.

10 Preferably, the magnesium dichloride is activated chemically. It can be accomplished by bringing a complex of magnesium dichloride in contact with the internal electron donor and the titanium tetrachloride, whereby the complex is converted to activated magnesium dichloride supporting the electron donor and the titanium tetrachloride.

15 According to a preferred embodiment of the invention, said complex of magnesium dichloride is a solid adduct of magnesium dichloride and an alcohol, which solid adduct has the formula (1)



wherein n is 1-6 and R is a C₁-C₁₀-alkyl. Preferably, n is 2-4 and R is a C₁-C₃-alkyl.
20 Most preferably, the solid adduct of magnesium dichloride and an alcohol having the formula (1) is the complex MgCl₂·3C₂H₅OH.

The solid adduct of magnesium dichloride and alcohol having the formula (1) is conveniently prepared by heating and melting the magnesium dichloride and the alcohol together, dispersing or spraying the melt into small droplets and solidifying
25 the droplets by contact with a cooled medium.

The dispersion of the melt into small droplets may typically take place by pouring the melt into hot silicon oil under stirring, forming a hot dispersion of molten droplets in silicon oil. Then, the solidification is brought about by pouring the hot dispersion into cold liquid hydrocarbon.

30 Preferably, the melt is sprayed by means of pressurized inert gas through a die into a space containing cold inert gas, whereby the small droplets are formed and solidified very rapidly. This process is called spray crystallization.

Finally, said solid adduct in powder form is contacted with the titanium tetrachloride, part of which removes alcohol and exposes activated sites on the surface of the magnesium dichloride, and part of which coordinates to the activated sites, as well as with the internal electron donor, which also coordinates to the
5 activated sites. The result is an active catalyst component comprising magnesium dichloride supporting coordinated electron donor and titanium tetrachloride.

As the internal electron donor to be contacted, any organic compound which contains an electron donating atom such as N, P, O and S, gives catalytic activity and enables stereospecific polymerization, can be selected. The art knows a
10 multitude of suitable electron donors for this purpose. Preferably, the internal electron donor is a C₁-C₁₄ alkyl ester of a carboxylic acid. Typical such esters are C₁-C₁₄-alkyl esters of aliphatic dicarboxylic acids such as maleic acid, malonic acid and cyclohexanedicarboxylic acid, C₁-C₁₄-alkyl esters of aromatic monocarboxylic acids such as substituted and unsubstituted benzoic acids, and C₁-C₁₄-alkyl esters of
15 aromatic dicarboxylic acids, preferably phthalic acid.

According to a preferred embodiment of the invention, the internal electron donor to be contacted is a C₄-C₁₄ alkyl ester of a carboxylic acid such as a C₄-C₁₄ alkyl ester of an aromatic carboxylic acid. More preferably, the internal electron donor is a di-C₄-C₁₄-alkyl ester of a dicarboxylic acid. Most preferably, the internal electron
20 donor is a di-C₄-C₁₄-alkyl phthalate such as dioctyl phthalate DOP.

The first organoaluminium compound of the invention preferably has the formula (2):



wherein R is a C₁-C₁₂ alkyl, X is a halogen, m is 1 or 2 and $0 \leq n \leq (3m-1)$.
25 Preferably, it is a trialkyl aluminium and most preferably, it is triethyl aluminium TEA. The second organoaluminium compound may be chosen freely among any organoaluminium compounds acting as cocatalysts. However, the second organoaluminium compound is preferably the same as said first organoaluminium compound.

30 In the invention, the first reaction mixture may be contacted with said second organoaluminium cocatalyst and an external electron donor. Electron donors which have proved to be highly stereospecific *per se* are chemically too unstable to be added directly to the magnesium dichloride/titanium tetrachloride reaction step. Therefore, it has been found, that it is advantageous to add an internal electron

donor (see above) to occupy sites on the MgCl_2 and then replace it by an external electron donor.

The external donor is different from the internal donor and is preferably selected from hydrocarbyloxy silane compounds and hydrocarbyloxy alkane compounds.

5 Typical hydrocarbyloxy silane compounds have the formula (3)



wherein R'' is an α - or β -branched C_3 - C_{12} -hydrocarbyl, R''' a C_1 - C_{12} -hydrocarbyl, and n is an integer 1-3.

10 More specific examples of the hydrocarbyloxy silane compounds which are useful as external electron donors in the invention are diphenyldimethoxy silane, dicyclopentyldimethoxy silane, dicyclopentyldiethoxy silane, cyclopentylmethyldimethoxy silane, cyclopentylmethyldiethoxy silane, dicyclohexyldimethoxy silane, dicyclohexyldiethoxy silane, cyclohexylmethyldimethoxy silane, cyclohexylmethyldiethoxy silane, methylphenyldimethoxy silane, diphenyldiethoxy silane, cyclopentyl-
15 trimethoxy silane, phenyltrimethoxy silane, cyclopentyltriethoxy silane, phenyltriethoxy silane.

Most preferably, the alkoxy silane compound having the formula (3) is dicyclopentyl dimethoxy silane or cyclohexylmethyl dimethoxy silane.

The activation step described above is followed by a prepolymerization.

20 In the prepolymerization, a premonomer is prepolymerized in the presence of at least said second reaction mixture to give a prepolymerizate. Preferably, the atomic ratio $\text{Al}_{1+2}/\text{Tr}$ between, on one hand, the aluminium of the first organoaluminium compound and the second organoaluminium compound taken together, and, on the other hand, the transition metal (Tr), is from about 1 to about 10, most preferably
25 from about 3 to about 8.

In the prepolymerization, the amount of said olefin premonomer is preferably such that the obtained weight ratio between the prepolymer obtained therefrom and said solid transition metal compound is between 1 and 10, most preferably between 1 and 5.

30 The used olefin premonomer may be any suitable olefin, but preferably it is a C_2 - C_6 olefin, more preferably ethene or propene, most preferably ethene. The prepoly-

merization is preferably carried out at a temperature between about 5 °C and about 40 °C, most preferably at a temperature between about 10 °C and about 30 °C.

In the invention, the active components are treated with a wax, fat, solid paraffin or the like. Preferably, the invention uses a wax. A wax is a natural or artificial solid substance, which is kneadable or plastic at 20 °C and melts above 40 °C without decomposing, exhibiting viscous behaviour or being drawable into a thread, and which in its solid state is polishable under light pressure. Preferably the wax used in the invention is a low molecular weight and/or atactic olefin polymer, preferably a low molecular weight (less than ca 10,000 g/mol) polyethylene wax or an atactic polypropylene wax.

A fat is a spreadable, solid or semi-solid viscous biogenic product, which consists essentially of mixed glycerine esters of higher fatty acids having an even number of carbon atoms. A solid paraffin is a saturated aliphatic hydrocarbon or a mixture of such hydrocarbons having a low tendency towards forming chemical bonds and having a solidification point of between 50 °C and 62 °C.

In the invention, the solid transition metal component and the organoaluminium cocatalyst are contacted in the presence of an oil. An oil is a water insoluble substance which is liquid at room temperature and has a relatively low vapor pressure, such as a mineral oil, synthetic oil or a biogenic oil. In the invention a synthetic oil obtained by oligomerizing decene, fractionating the product to an average of 30 carbon atoms, and hydrogenating is preferable. Such a product is the 4 cSt PAO (polyalphaolefin) oil of Neste Oy. Other suitable oils are e.g. the mineral oil Technol 68, the oils Fina Vesta A 180B and Shell Ondina Oil 68 as well as the white oil Technol 68. The oligomer distribution thereof is 85% C₃₀, 13% C₄₀ and 2% C₅₀. Silicon oil is not recommended for the purpose of the invention.

In addition to the above described catalyst system for the polymerization of olefins, the invention relates to the above described process for preparing a catalyst system of said type.

Further, the invention relates to a process for the polymerization of an olefin, in which an olefin is contacted with a catalyst system of the above described type. The result is an improved polyolefin having low catalyst residue, good processability and a low fraction of fines. Preferably, said olefin is a C₃-C₆- α -olefin or a mixture thereof, more preferably propene or a mixture of propene and less than 20% by weight of ethene.

The catalyst system according to the invention may be used as such for the polymerization of said olefin. This means that essentially all of the organoaluminium compound(s) has been added during the two or more activation steps. Alternatively, only a part of the total amount of organoaluminium compound has been added during the activation, whereby a third organoaluminium compound is added in connection with the contacting of the olefin and the claimed catalyst system. The third organoaluminium compound is preferably the same as said first and/or second organoaluminium compound. In the claimed polymerization process, the atomic ratio Al/Tr, preferably Al/Ti is preferably 40-1000, most preferably about 50 to about 500.

In the polymerization process of the invention, the molar mass of the polyolefin may be regulated by using a chain transfer agent like hydrogen. Preferably, the hydrogen is contacted with said catalyst system and said olefin under polymerization conditions, preferably in an amount giving propylene polymer having a melt flow rate MFR₂ of between 0.03 g/10 min and 2000 g/10 min, more preferably 0.3-1000 g/10 min, most preferably between 1.0 g/10 min and 400 g/10 min.

Examples

Example 1

Highly active catalyst and highly active polymerization conditions were used to test the properties of the novel system.

Catalyst handling, preactivation

A propylene polymerization catalyst of Ziegler-Natta-type (according to Finnish Patent No. 88047) was preactivated with a low amount of triethylaluminium (TEA) in 100 dm³ pilot scale reactor. In catalyst preactivation (500 g) dry catalyst was at first fed into polyalphaolefin, PAO 4 st (31 dm³) (manufactured by Neste Oyj) oil at 30 °C temperature. Oil/catalyst mixture was cooled down to 10 °C and TEA having Al/Ti molar ratio of 1.5 was mixed in (The titanium content of the catalyst was 2.0 wt-%). After 1 hour mixing, temperature was increased and 18 dm³ grease, White Protopet (manufactured by Witco) was added at 40 °C to keep the viscosity moderate. The mixture was cooled down to room temperature before use in polymerization. The catalyst concentration in oil-grease mixture was 10 g/dm³.

Catalyst feed and prepolymerization

Said mixture of catalyst and viscous medium was fed with non valve piston pump according to Finnish patent no 94164. The catalyst was contacted with triethylaluminium (TEA) and dicyclopentyldimethoxysilane (DCPDMS) in pipeline. Al/Ti molar ratio was 250 mol/mol and Al/D molar ratio was 10. The contact time between catalyst, cocatalyst and donor was 15 seconds.

The catalyst was flushed with propylene (15 kg/h) to the prepolymerization reactor (CCSTR = Compartmented Continuous Stirred Tank Reactor) in which also TEA and DCPDMS were fed. The CCSTR reactor has been introduced in Finnish Patent application no 961152. The prepolymerization reactor was operated at 55 barg pressure and at the temperature of 30 °C.

The mean residence time of the catalyst was 8 minutes in prepolymerization reactor. TEA/Ti molar ratio was kept at 250 and molar ratio was kept at 10.

Polymerization conditions

Polymerization conditions in loop reactor during the test were as follows: reaction temperature 70 °C, pressure 40 barg polymer residence time 1.5 hours. The MFR measured according to ISO 1133 (2.16 kg, 230 °C) of the produced PP-homopolymer was controlled to be 9-12 via hydrogen feed. The test was duplicated in example 1b. Product characteristics are shown in Table 1.

The xylene soluble fraction (XS) fraction was measured and calculated as follows:

2.0 g of polymer are dissolved in 250 ml p-xylene at 135 °C under agitation. After 30 ± 2 minutes the solution is allowed to cool for 15 minutes at ambient temperature and then allowed to settle for 30 minutes at 25 ± 0.5 °C. The solution is filtered with filter paper into two 100 ml flasks.

The solution from the first 100 ml vessel is evaporated in nitrogen flow and the residue is dried under vacuum at 90 °C until constant weight is reached.

$$XS\% = (100 \times m_1 \times v_0) / (m_0 \times v_1)$$

m_0 = initial polymer amount (g)

m_1 = weight of residue (g)

v_0 = initial volume (ml)

v_1 = volume of analyzed sample (ml)

PS in the average particle size according to ASTM 1921. The tensile modulus was determined according to ISO 527 (1 mm/1 min). The activity has been calculated from the added amount of catalyst and the amount of product recovered per hour (kg PP/h/g catalyst/h).

5 **Example 2**

The procedure was the same as in example 1 but cocatalyst and donor split according to Finnish patent application no FI 952175 was used in this test. 10 wt-% of the total amount of TEA and 10 wt-% of total amount of DCPDMS were fed to catalyst activation and the rest of the TEA and DCPDMS were fed directly to the
10 polymerization.

The test was duplicated in example 2b. Product characteristics are shown in Table 1.

Comparative Example 3

The procedure was the same but preactivation was not carried out for this reference
15 test. Highly active catalyst (according to Finnish Patent no 88047) and polymerization system were used to test the properties of conventional system.

Catalyst handling

A propylene polymerization catalyst of Ziegler-Natta-type was mixed with an oil-grease mixture. A mixture of synthetic oil (polyalphaolefin, PAO 4 st) and grease
20 (White Protopet) was made separately having weight ratio of 1.75/1 and total amount of 50 dm³. The dry catalyst (500 g) was fed slowly into the reactor and the mixture was mixed half an hour at the temperature of 70 °C and the reactor was cooled down to room temperature. The catalyst concentration in oil-grease mixture was 10 g/dm³.

Catalyst feed and prepolymerization, polymerization

The catalyst was fed, prepolymerized and polymerized as in example 1. The test was duplicated in example 3b. Product characteristics are shown in Table 1.

Table 1

Example	Catalyst preact	Al, Don split	Fines <0.25 mm	Av. PS mm	XS wt-%	BD g/cm ³	Act. kg/g cat.	MFR ₂ g/10 min.	Tens. Mod. MPa
1a	yes	no	9.3	1.24	1.8	0.4	44.3	12	nm
1b	yes	no	10.7	1.192	1.9	0.39	44.3	11	1720
2a	yes	yes	12.0	1.192	1.90	0.42	48.6	13	nm
2b	yes	yes	10.6	1.212	1.90	0.42	47.1	12	1720
3a	no	no	25.4	0.791	1.80	0.40	33.5	9	nm
3b	no	no	22.7	0.772	1.60	0.40	33.8	9	1710

5

nm = not measured

Claims

1. A catalyst system for the polymerization of α -olefins, the catalyst being prepared by a process including a catalyst activation comprising the contacting of a solid transition metal compound with an organoaluminium compound, and a catalyst
5 prepolymerization comprising the polymerization of a premonomer in the presence of the activated catalyst, **characterized** in that the catalyst activation comprises a first step of contacting the solid transition metal compound with a first organoaluminium compound in the presence of an oil to give a first reaction mixture, and a second step of contacting the first reaction mixture with a second organoaluminium
10 compound to give a second reaction mixture, the second organoaluminium compound being the same as or different from the first organoaluminium compound.
2. A catalyst system according to claim 1, **characterized** in that in said first step, a mixture consisting essentially of said solid transition metal catalyst component and said oil is preactivated with said first organoaluminium compound.
- 15 3. A catalyst system according to claim 1 or 2, **characterized** in that in said first step, the weight ratio between said solid transition metal compound and said oil is between 0.1 and 5, preferably between 0.2 and 1, most preferably between 0.3 and 0.8.
4. A catalyst system according to claim 2 or 3, **characterized** in that in said first
20 step, said mixture consisting essentially of said solid transition metal compound and said oil has been prepared by heating them together at an elevated temperature, preferably at a temperature between about 26 °C and about 100 °C, most preferably at a temperature between about 30 °C and about 80 °C.
5. A catalyst system according to any preceding claim, **characterized** in that in
25 said first step, said solid transition metal compound, said organoaluminium compound and said oil are precontacted at a lowered temperature, preferably at a temperature between about -20 °C and about +20 °C, most preferably at a temperature between about 0 °C and about +16 °C.
6. A catalyst system according to any preceding claim, **characterized** in that in
30 said first step, said organoaluminium compound (Al_1) and said solid transition metal (Tr) compound are contacted in the presence of said at least a part of the oil in an atomic ratio Al_1/Tr of between about 0.5 and about 5, preferably between about 1 and about 3.

7. A catalyst system according any preceding to claim, characterized in that in said first step, said first reaction mixture is further contacted with a wax, fat, solid paraffin or the like to give a waxed first reaction mixture.

5 8. A catalyst system according to claim 7, characterized in that in said first step, said wax, fat, solid paraffin or the like is added at higher temperature than its melting point.

9. A catalyst system according to any preceding claim, characterized in that in said second step, said first reaction mixture or said waxed first reaction mixture is further activated with said second organoaluminium compound.

10 10. A catalyst system according to claim 9, characterized in that in said second step, said first reaction mixture or said waxed first reaction mixture is contacted with an external electron donor.

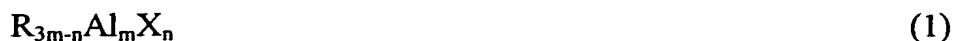
11. A catalyst system according to any of claims 7 to 10, characterized in that the weight ratio between the total amount of said oil and the total amount of said wax, fat, solid paraffin or the like is such that the viscosity of their mixture at 20-25 °C is about 1 Pa·s to about 15 Pa·s, preferably about 4 Pa·s to about 10 Pa·s.

12. A catalyst system according to any preceding claim, characterized in that the atomic ratio between the aluminium (Al₁) of said first organoaluminium compound and the aluminium (Al₂) of said second organoaluminium compound Al₁/Al₂ is between about 0.001 and about 1, preferably between about 0.01 and about 0.1.

13. A catalyst system according to any preceding claim, characterized in that the atomic ratio between the aluminium (Al) of the total amount of organoaluminium compound and the transition metal (Tr) of the solid transition metal compound Al/Tr is between about 10 and about 1000, preferably between about 50 and about 500.

14. A catalyst system according to any preceding claim, characterized in that said solid transition metal compound has been prepared by contacting at least magnesium dichloride or a complex thereof, titanium tetrachloride and an internal electron donor.

15. A catalyst system according to any preceding claim, characterized in that said first organoaluminium compound has the formula (1):



wherein R is a C₁-C₁₂ alkyl, X is a halogen, m is 1 or 2 and 0 ≤ n ≤ (3m-1), and preferably is a trialkyl aluminium, most preferably triethyl aluminium TEA.

16. A catalyst system according to any preceding claim, characterized in that said second organoaluminium compound is the same as said first organoaluminium compound.

17. A catalyst system according to any preceding claims, characterized in that in the prepolymerization, the premonomer is polymerized in the presence of at least said second reaction mixture to give a prepolymerizate.

18. A catalyst system according to any preceding claim, characterized in that in the prepolymerization, the atomic ratio Al₁₊₂/Tr between, on one hand, the aluminium (Al₂) of said second organoaluminium compound and the aluminium (Al₁) of first organoaluminium compound taken together, and, on the other hand, the transition metal (Tr) if said solid transition metal compound, is from about 1 to about 10, preferably from about 3 to about 8.

19. A catalyst system according to any preceding claim, characterized in that in the prepolymerization, the amount of said olefin premonomer is such that the obtained weight ratio between the prepolymer obtained therefrom and said solid transition metal catalyst compound is between 1 and 10, preferably between 1 and 5.

20. A catalyst system according to any preceding claim, characterized in that in the prepolymerization, said olefin premonomer is ethene.

21. A process for the polymerization of an olefin, characterized in that an α-olefin is contacted with a catalyst system according to any of claims 1-20.

22. A process according to claim 21, characterized in that said α-olefin is a C₃-C₆-α-olefin or a mixture thereof, preferably propene or a mixture of propene and less than 20% by weight of ethene.

23. A process according to claim 21 or 22, characterized in that said α-olefin is copolymerized with another α-olefin monomer or ethene.

24. A process according to claim 21, 22 or 23, characterized in that the olefin is contacted with a third organoaluminium compound.

25. A process according to claim 24, **characterized** in that the third organo-aluminium compound is the same as said first and/or second organoaluminium compound.

5 26. A process according to claim 24 or 25, **characterized** in that the total amount of aluminium (Al) is such that the atomic ratio Al/Tr is 40-1000, preferably about 50 to about 500.

10 27. A process according to any of claims 21 to 26, **characterized** in that hydrogen is contacted with said catalyst system and said olefin under polymerization conditions, preferably in an amount giving propylene polymer having a melt flow rate MFR₂ of between 0.3 g/10 min and 2000 g/10 min, more preferably 0.3-1000 g/10 min, most preferably between 1.0 g/10 min and 400 g/10 min.

TENT COOPERATION TREATY

PCT

REC'D 24 APR 2001

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)



Applicant's or agent's file reference 49513	FOR FURTHER ACTION See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/FI00/00098	International filing date (day/month/year) 11/02/2000	Priority date (day/month/year) 12/02/1999
International Patent Classification (IPC) or national classification and IPC C08F4/44		
Applicant BOREALIS TECHNOLOGY OY		

- This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.
- This REPORT consists of a total of 5 sheets, including this cover sheet.
 - ☐ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of sheets.

- This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☐ Certain defects in the international application
- VIII ☒ Certain observations on the international application

Date of submission of the demand 05/09/2000	Date of completion of this report 20.04.2001
Name and mailing address of the international preliminary examining authority:  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer Adams, F Telephone No. +49 89 2399 8511 

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/FI00/00098

I. Basis of the report

1. With regard to the **elements** of the international application (*Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17)*):

Description, pages:

1-15 as originally filed

Claims, No.:

1-27 as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☐ the claims, Nos.:
- ☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/FI00/00098

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims	1-27
	No:	Claims	
Inventive step (IS)	Yes:	Claims	1-27
	No:	Claims	
Industrial applicability (IA)	Yes:	Claims	1-27
	No:	Claims	

2. Citations and explanations see separate sheet

VIII. Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:
see separate sheet

Ad V:

- 1). The present application satisfies the criteria set forth in Article 33(2) and 33(3) PCT because the subject-matter of the claims is new and inventive in respect of prior art as defined in the regulations (Rule 64(1)-(3) PCT).

None of the cited documents discloses a catalyst system for the polymerization of alpha-olefins which is prepared by a first activation step comprising contacting a solid transition metal compound with an organoaluminum compound in the presence of an oil and contacting in a second step the obtained reaction mixture with an organoaluminum compound to obtain an activated catalyst, and a prepolymerization step comprising prepolymerizing a monomer in the presence of the activated catalyst, or makes such a catalyst system obvious.

- 2). EP-A-0588277 discloses the activation of a Ziegler-type catalyst by contacting the catalyst with TEAL in the presence of hexane and the prepolymerization of the activated catalyst with propene. The activated and prepolymerized catalyst is subsequently diluted with a mineral oil and used for propene polymerization in the presence of further TEAL. Nothing is said about the activation in the presence of mineral oil prior to the prepolymerization.

EP-A-0887379 discloses the polymerization of propene in the presence of a prepolymerized transition metal catalyst and an organoaluminum compound as an activating agent. The prepolymerization may be carried out in the presence of a viscous substance such as an olefinic wax. Nothing is said about an activation step in the presence of an oil.

US-A-5641721 (D1) discloses the activation of a titanium based catalyst with triethylaluminum in the presence of PAO oil and the prepolymerization of the thus obtained activated catalyst with propylene in the presence of further triethyl aluminum (cf. example 1; column 4, lines 39 to column 5, lines 54). The difference between the catalyst system as claimed and the catalyst system of D2 is that in D2 the procatalyst (i.e. the transition metal) and the cocatalyst, (i.e. the organoaluminum compound), are first combined and then added to the oil (see column 5, lines 18-22; column 7, lines 31-37).

EP-A-0573862 discloses the preparation of a supported titanium-based catalyst prepared by heating MgCl_2 , TiCl_4 and vaseline oil, the activation of the supported catalyst with TEAL and the subsequent prepolymerization of the catalyst with propene. Nothing is said about an activation step in the presence of an oil and TEAL.

Ad VIII:

- 1). The definition of the catalyst system comprising a "solid transition metal compound" is unclear (Art. 6 PCT). It appears from the description on page 5, line 21 and the examples that the transition metal of the invention is titanium. Alternatively, defining the transition metal as a metal belonging to the Groups 4 to 6 of the Periodic Table of Elements (page 1, lines 28-32) may also clarify claim 1.
- 2). The term "oil" used in claim 1 is vague and unclear and leaves the reader in doubt as to the meaning of the technical feature to which it refers, thereby rendering the definition of the subject-matter of said claim unclear (Article 6 PCT). It should be noted that it depends inter alia on the temperature whether a compound is an oil or a wax.
- 3). Unclear expressions like "about" and "and the like" were not deleted throughout the claims and the description (Art. 6 PCT).

PCT REQUEST

49513

Original (for SUBMISSION) - printed on 11.02.2000 09:57:08 AM

0	For receiving Office use only	
0-1	International Application No.	
0-2	International Filing Date	
0-3	Name of receiving Office and "PCT International Application"	
0-4	Form - PCT/RO/101 PCT Request	
0-4-1	Prepared using	PCT-EASY Version 2.90 (updated 15.12.1999)
0-5	Petition The undersigned requests that the present international application be processed according to the Patent Cooperation Treaty	
0-6	Receiving Office (specified by the applicant)	National Board of Patents and Registration (Finland) (RO/FI)
0-7	Applicant's or agent's file reference	49513
I	Title of invention	AN ALFA-OLEFIN POLYMERIZATION CATALYST SYSTEM AND ITS USE FOR THE POLYMERIZATION OF ALFA-OLEFINS
II	Applicant	
II-1	This person is:	applicant only
II-2	Applicant for	all designated States except US
II-4	Name	BOREALIS A/S
II-5	Address:	Lyngby Hovedgade 96 DK-2800 Lyngby Denmark
II-6	State of nationality	DK
II-7	State of residence	DK
III-1	Applicant and/or inventor	
III-1-1	This person is:	applicant and inventor
III-1-2	Applicant for	US only
III-1-4	Name (LAST, First)	ALASTALO, Kauno
III-1-5	Address:	Karjapolku 4 FIN-06400 Porvoo Finland
III-1-6	State of nationality	FI
III-1-7	State of residence	FI

PCT REQUEST

49513

Original (for SUBMISSION) - printed on 11.02.2000 09:57:08 AM

III-2	Applicant and/or inventor	
III-2-1	This person is:	applicant and inventor
III-2-2	Applicant f r	US only
III-2-4	Name (LAST, First)	LESKINEN, Pauli
III-2-5	Address:	Kantakyläntie 6 D FIN-00640 Helsinki Finland
III-2-6	State of nationality	FI
III-2-7	State of residence	FI
IV-1	Agent or common representative; or address for correspondence The person identified below is hereby/has been appointed to act on behalf of the applicant(s) before the competent International Authorities as:	agent
IV-1-1	Name	BERGGREN OY AB
IV-1-2	Address:	P.O. Box 16 FIN-00101 Helsinki Finland
IV-1-3	Telephone No.	+358-9-693701
IV-1-4	Facsimile No.	+358-9-6933944
IV-1-5	e-mail	email.box@berggren.elisa.fi
V	Designation of States	
V-1	Regional Patent (other kinds of protection or treatment, if any, are specified between parentheses after the designation(s) concerned)	AP: GH GM KE LS MW SD SL SZ TZ UG ZW and any other State which is a Contracting State of the Harare Protocol and of the PCT EA: AM AZ BY KG KZ MD RU TJ TM and any other State which is a Contracting State of the Eurasian Patent Convention and of the PCT EP: AT BE CH&LI CY DE DK ES FI FR GB GR IE IT LU MC NL PT SE and any other State which is a Contracting State of the European Patent Convention and of the PCT OA: BF BJ CF CG CI CM GA GN GW ML MR NE SN TD TG and any other State which is a member State of OAPI and a Contracting State of the PCT
V-2	National Patent (other kinds of protection or treatment, if any, are specified between parentheses after the designation(s) concerned)	AE AL AM AT AU AZ BA BB BG BR BY CA CH&LI CN CR CU CZ DE DK DM EE ES FI GB GD GE GH GM HR HU ID IL IN IS JP KE KG KP KR KZ LC LK LR LS LT LU LV MA MD MG MK MN MW MX NO NZ PL PT RO RU SD SE SG SI SK SL TJ TM TR TT TZ UA UG US UZ VN YU ZA ZW

PCT REQUEST

49513

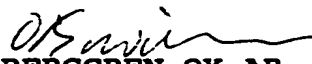
Original (for SUBMISSION) - printed on 11.02.2000 09:57:08 AM

V-5	Precautionary Designation Statement In addition to the designations made under items V-1, V-2 and V-3, the applicant also makes under Rule 4.9(b) all designations which would be permitted under the PCT except any designation(s) of the State(s) indicated under item V-6 below. The applicant declares that those additional designations are subject to confirmation and that any designation which is not confirmed before the expiration of 15 months from the priority date is to be regarded as withdrawn by the applicant at the expiration of that time limit.		
V-6	Exclusion(s) from precautionary designations	NONE	
VI-1	Priority claim of earlier national application		
VI-1-1	Filing date	12 February 1999 (12.02.1999)	
VI-1-2	Number	990283	
VI-1-3	Country	FI	
VI-2	Priority document request The receiving Office is requested to prepare and transmit to the International Bureau a certified copy of the earlier application(s) identified above as item(s):	VI-1	
VII-1	International Searching Authority Chosen	European Patent Office (EPO) (ISA/EP)	
VIII	Check list	number of sheets	electronic file(s) attached
VIII-1	Request	4	-
VIII-2	Description	15	-
VIII-3	Claims	4	-
VIII-4	Abstract	1	49513.txt
VIII-5	Drawings	0	-
VIII-7	TOTAL	24	
	Accompanying items	paper document(s) attached	electronic file(s) attached
VIII-8	Fee calculation sheet	✓	-
VIII-9	Separate signed power of attorney	✓	-
VIII-16	PCT-EASY diskette	-	diskette
VIII-17	Other (specified):	Copy of Official Action in FI 990283	-
VIII-18	Figure of the drawings which should accompany the abstract		
VIII-19	Language of filing of the international application	English	

PCT REQUEST

49513

Original (for SUBMISSION) - printed on 11.02.2000 09:57:08 AM

IX-1	Signature of applicant or agent	
IX-1-1	Name	BERGGREN OY AB
IX-1-2	Name of signatory	Olli-Pekka Saijonmaa
IX-1-3	Capacity	Patent Agent

FOR RECEIVING OFFICE USE ONLY

10-1	Date of actual receipt of the purported international application	
10-2	Drawings:	
10-2-1	Received	
10-2-2	Not received	
10-3	Corrected date of actual receipt due to later but timely received papers or drawings completing the purported international application	
10-4	Date of timely receipt of the required corrections under PCT Article 11(2)	
10-5	International Searching Authority	ISA/EP
10-6	Transmittal of search copy delayed until search fee is paid	

FOR INTERNATIONAL BUREAU USE ONLY

11-1	Date of receipt of the record copy by the International Bureau	
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PATENT COOPERATION TREATY

PCT

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference 49513	FOR FURTHER ACTION see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
International application No. PCT/ FI 00/ 00098	International filing date (day/month/year) 11/02/2000	(Earliest) Priority Date (day/month/year) 12/02/1999
Applicant BOREALIS A/S et al.		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 3 sheets.



It is also accompanied by a copy of each prior art document cited in this report.

1. Basis of the report

- a. With regard to the language, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.



the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

- b. With regard to any nucleotide and/or amino acid sequence disclosed in the international application, the international search was carried out on the basis of the sequence listing :



contained in the international application in written form.



filed together with the international application in computer readable form.



furnished subsequently to this Authority in written form.



furnished subsequently to this Authority in computer readable form.



the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.



the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished

2. ☐ Certain claims were found unsearchable (See Box I).

3. ☐ Unity of invention is lacking (see Box II).

4. With regard to the title,



the text is approved as submitted by the applicant.



the text has been established by this Authority to read as follows:

5. With regard to the abstract,



the text is approved as submitted by the applicant.



the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. The figure of the drawings to be published with the abstract is Figure No.



as suggested by the applicant.



because the applicant failed to suggest a figure.



because this figure better characterizes the invention.

☐ None of the figures.

INTERNATIONAL SEARCH REPORT

International Application No

PCT 00/00098

A. CLASSIFICATION OF SUBJECT MATTER
 IPC 7 C08F4/44 C08F10/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C08F

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 588 277 A (FINA TECHNOLOGY) 23 March 1994 (1994-03-23) page 2, line 46 - line 50 page 3, line 15 - line 20 page 5, line 50 -page 6, column 7 ---	1-27
A	EP 0 887 379 A (BOREALIS AS) 30 December 1998 (1998-12-30) column 12, line 23 - line 28 ---	1-27
A	US 5 641 721 A (PENTTI ISMO ET AL) 24 June 1997 (1997-06-24) claims, abstract column 4, line 53 - line 63 --- -/--	1-27

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search

7 June 2000

Date of mailing of the international search report

04.08.2000

Name and mailing address of the ISA

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 Fax: (+31-70) 340-3016

Authorized officer

HELENA DANIELSSON

INTERNATIONAL SEARCH REPORT

International Application No

PCT 00/00098

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	<p>EP 0 573 862 A (HIMONT INC) 15 December 1993 (1993-12-15) page 4, line 4 - line 16 page 6, line 25 - line 37 claims</p> <p>-----</p>	1-27

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT 00/00098

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP 0588277 A	23-03-1994	US 4767735 A AT 103614 T CA 1274945 A DE 3789500 D DE 3789500 T EP 0279153 A ES 2050670 T JP 2771169 B JP 63270706 A JP 2905184 B JP 10182739 A JP 2905185 B JP 10182740 A KR 9508153 B US 5122583 A	30-08-1988 15-04-1994 02-10-1990 05-05-1994 04-08-1994 24-08-1988 01-06-1994 02-07-1998 08-11-1988 14-06-1999 07-07-1998 14-06-1999 07-07-1998 25-07-1995 16-06-1992
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US 5641721 A	24-06-1997	FI 925913 A US 6040260 A DE 69318386 D DE 69318386 T EP 0607703 A NO 934863 A US 5733989 A	30-06-1994 21-03-2000 10-06-1998 03-12-1998 27-07-1994 30-06-1994 31-03-1998
EP 0573862 A	15-12-1993	IT 1260497 B AT 164380 T AU 658478 B AU 3311593 A CA 2092639 A CN 1079968 A, B DE 69317599 D DE 69317599 T ES 2114967 T FI 932442 A JP 6093034 A MX 9301698 A NO 931933 A RU 2081132 C	09-04-1996 15-04-1998 13-04-1995 02-12-1993 30-11-1993 29-12-1993 30-04-1998 24-09-1998 16-06-1998 30-11-1993 05-04-1994 01-11-1993 30-11-1993 10-06-1997